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Thin epitaxial Al and Cu films grown on CaF_2/Si(111)

N. L. Yakovlev†, Y. V. Shusterman‡ and L. J. Schowalter‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Department of Physics, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

Abstract. Molecular beam epitaxy was used to grow single crystal CaF_2, Al and Cu films on Si(111). Reflection high energy electron diffraction indicated that Al film was epitaxial when it was grown on CaF_2/Si, and that epitaxial Cu film can be grown on Al/CaF_2/Si heteroepitaxial substrates. Room temperature measurements of resistivity of Al films 10 to 300 nm thick agree with the Fuchs-Sondheimer model, in which only diffuse scattering of conduction electrons occurs at the film interfaces. For 50 to 1000 nm thick Cu films, the resistivity size effect is greater than the prediction of this model.

Introduction

In the next decade, the width of some metal lines inside integrated circuits will decrease to approximately 50 nm, which is comparable to the mean free path of conduction electrons in both Al and Cu at room temperature. At those small dimensions, electron scattering from metal surfaces will play an important role in electron transport. The well-established way to characterize surface scattering is to study the electrical properties of thin metal films. Most previous work related to Al and Cu has been done on polycrystalline films deposited on SiO_2 [1, 2]. However, in such cases, the scattering at grain boundaries may be dominant. To avoid this masking effect, it is desirable to work with single-crystal films.

The epitaxial CaF_2 on Si is a convenient choice for a single-crystal insulating substrate [3]. Epitaxy has been reported previously for both Al and Cu on CaF_2(111) [4, 5], but the initial stages of their growth have not been addressed. In this work, we have studied thin Al and Cu films grown on CaF_2/Si(111) by molecular beam epitaxy (MBE). The samples were characterized by reflection high-energy electron diffraction (RHEED), scanning tunnelling microscopy (STM) and atomic force microscopy (AFM). We have also measured the resistivity dependence on film thickness for both Al and Cu, fitted these data to the Fuchs-Sondheimer size effect model, and discussed the relationship between resistivity and structural properties.

Growth

The films studied in this work were grown in a commercial VG90S MBE apparatus with a base pressure of 3×10^{-8} Pa. Molecular beam of CaF_2 was produced by sublimation of the fluoride from boron nitride crucible. The Al and Cu were deposited from electron beam evaporators. The RHEED patterns were obtained at electron energy of 13 keV and beam incidence angle of 2.5°. The STM work was performed on a locally designed instrument coupled to the MBE chamber. The AFM images were obtained by a Digital Instruments "Dimension 3100" microscope in the tapping mode with a tip having the apex radius below 30 nm.

When Al is deposited on traditional SiO_2, the film starts growing as isolated clusters, Fig. 1(a). RHEED showed that their crystallographic orientation is random. At room

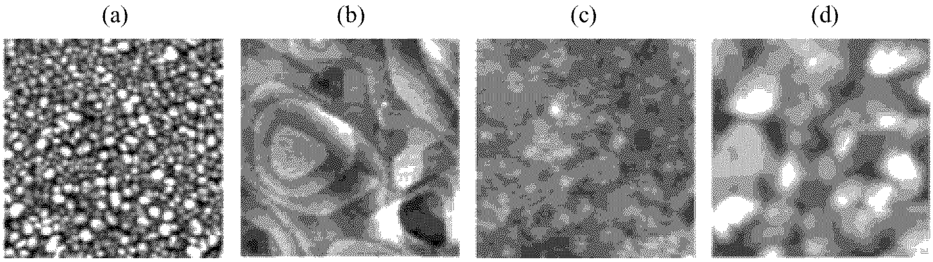

Fig. 1. AFM images of (a) 10 nm of Al on SiO_2, (b) 50 nm of CaF_2 on Si(111), (c) 10 nm of Al on CaF_2, (d) 130nm of Cu on Al seed layer. Scan size is 500 nm, grey scale is 8 nm in (a), 2 nm in (b) and (c), 5 nm in (d). Root-mean-square roughness is 5 nm in (a), 0.5 nm in (b), 0.3 nm in (c), 3.5 nm in (d).

temperature and relatively high deposition rate of 1 nm/s, a continuous film can be obtained when average thickness of Al is around 10 nm.

Epitaxial films were grown on hydrogen-terminated Si(111) substrates prepared by wet etching in 40% NH_4F. One molecular layer (ML) of CaF_2 was deposited on the surface at 250°C, then heated to 770°C and the CaF_2 deposition was continued. In this way, we obtained a well-reacted CaF_2/Si(111) interface without long exposure of the bare Si surface to an imperfect vacuum (the pressure rose to 10^{-6} Pa when the substrate was at high temperature). The surface of 50 nm thick layer had atomically flat terraces 50 nm wide, as shown in Fig. 1(b).

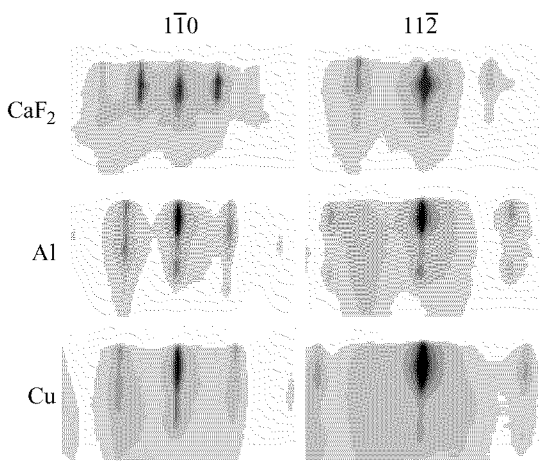

Fig. 2. RHEED patterns showing epitaxy of Cu on Al on CaF_2(111).

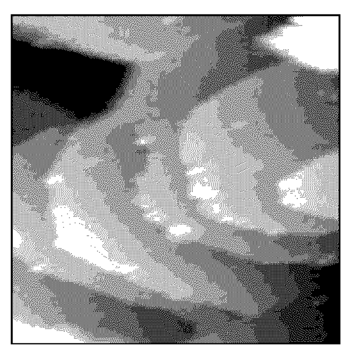

Fig. 3. STM image of 10 nm Al on CaF_2; scan 20 nm, grey scale 0.8 nm.

Al film grown on this surface was epitaxial, as shown by RHEED images in Fig. 2. The streak pattern was consistent with that from (111) face of bulk Al. To improve continuity of very thin films, we increased nucleation density as follows. Additional 1.5 ML of CaF_2 was grown at room temperature, as suggested in Ref. [6], this created high concentration of atomic steps on the CaF_2 surface. The metal layer was grown also at room temperature at high deposition rates above 1 nm/s. The resulting rms roughness was below 0.3 nm on a 500 nm AFM scan, Fig. 1(c). On STM images of this surface shown in Fig. 3, one can see individual monolayer steps; some of them originate at outlets of screw dislocations.

They are due to large lattice mismatch between Al and CaF_2 (the ratio of lattice constants is 1.36), and their density was estimated to be $5 \times 10^{11}/cm^2$.

For Cu deposition on CaF_2, RHEED showed the formation of a polycrystalline film. However, we succeeded in growing epitaxial Cu by depositing it on a 1 nm-thick Al seed layer grown on CaF_2. Only Cu(111) streaks were visible in RHEED, with a spacing indicative of the bulk Cu lattice spacing, as shown Fig. 2. This result is consistent with the report of Cu epitaxy on the (111) surface of bulk Al [7]. During the growth at room temperature, Cu surface became rough quickly, so after the growth of 50 nm, the temperature was increased to 100°C and the films were grown up to intended thickness. The rms roughness of the final surface was below 5% of the average film thickness, Fig. 1(d).

Resistivity measurements

The insulator covered the substrate uniformly. The metals were deposited onto it through a shadow mask designed to measure sheet resistance ex-situ by a linear 4-point probe, as described in ref. [8]. Precision of the resistance measurements was better than 0.1%. For films thicker than 50 nm, the thickness was measured with precision of ±2 nm using Tencor Alpha-Step 200 profilometer. For thinner films, the thickness was obtained with a precision of ±0.5 nm from AFM scan at the metal shadow edge. Oxidation of Al films was taken into account like it was done in ref. [8].

The thin-film resistivity data for epitaxial Al and Cu on CaF_2 are presented in Fig. 4, along with the fit by the Fuchs-Sondheimer model [9], which treats additional contribution to the resistivity arising from diffuse scattering of electrons at the film surfaces. Even though this model is very crude and has been refined several times (see ref. [10] for review), it is still customarily used for analysis. It expresses the resistivity as

$$\rho(h) = \rho_\infty \left(1 + \frac{C}{h}\right) \quad \text{with} \quad C = \frac{3}{8}\lambda(1 - p),$$

where ρ is the resistivity of the film, ρ_∞ is that of the bulk, h is the metal thickness, λ is the bulk electron mean free path, p is the probability of a specular surface scattering event. We cast the equation into such a form because C is what is actually determined from the fit to the experimental data; thus we obtained $C = 6.4 \pm 0.6$ nm for Al and $C = 26 \pm 2$ nm for Cu.

For the surface scattering contribution, it is expected that $p > 0$ and hence $C < 3\lambda/8$. Here, λ is the product of Fermi velocity and relaxation time. Taking those from ref. [11],

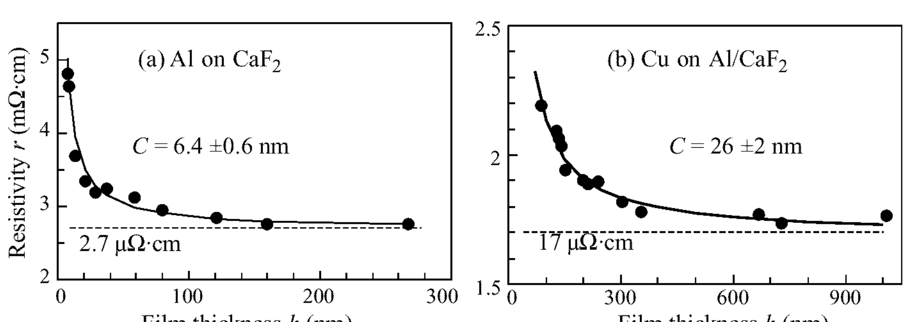

Fig. 4. Resistivity size effect for (a) Al and (b) Cu epitaxial films on CaF_2. The dotted line is the bulk resistivity $2.7\mu\Omega \cdot$ cm for Al and $1.7\mu\Omega \cdot$ cm for Cu.

we obtain $\lambda = 16$ nm for Al and $\lambda = 42$ nm for Cu. With these values, $3\lambda/8 = 6$ nm for Al, so we can say that $C \approx 3\lambda/8$ within the experimental precision and that $p \approx 0$, i.e. almost all of the surface scattering events are diffuse. This is not surprising since both Al interfaces with CaF_2 and the oxide are expected to be atomically rough and hence should efficiently scatter conduction electrons whose Fermi wavelength is comparable with the lattice constant.

However for Cu, $3\lambda/8 \approx 16$ nm and $C > 3\lambda/8$. So we conclude that in addition to diffuse surface scattering, another contribution to the resistivity size effect must be present which is also inversely proportional to the thickness. Analysis of RHEED images with variation of electron beam azimuth showed that there are domains with small in-plane misorientation $\pm 10^\circ$. Scattering at their boundaries can cause additional resistivity of thin films. In thicker films, these domains are overgrown by exactly epitaxial Cu, so relative contribution of this effect is smaller.

To conclude, thin epitaxial Al films on $CaF_2(111)$ can be grown very smooth. Their crystal quality is good enough to perform meaningful studies of the resistivity size effect due to surface scattering of electrons, without the masking effect of grain boundary scattering. These Al films can also be used as a template to grow other metals epitaxially, notably Cu.

Acknowledgements

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